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Density and comparative refractive index study on mixing properties of binary liquid mixtures of oleic acid and alcohols

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ABSTRACT

Density and refractive index have been determined for binary liquid mixture of oleic acid with alcohols like methanol, ethanol, n-propanol, n-butanol and iso-butanol at 303.15K, 308.15K and 313.15K. For comparative nine relations Lorentz -Lorenz (L-L), Weiner (W), Heller (H), Gladstone-Dale (G-D), Arago-Biot (A-B), Eykman (Eyk), Newton (Nw), Eyring-John (EJ), Oster (Os) has been used for determining the refractive index of different liquid mixtures. From the experimentally measured values, refractive index deviation at different temperatures have been computed and fitted to the Redlich- Kister polynominal equation to derive the binary coefficient and standard deviation. Comparison of various mixing rules has been expressed in term of average percentage deviation. All nine theoretical mixing rules performed well within the experimental error. Weiner equation has proved to be better then other relation. n^E values are positive over whale concentration range in case of all binary mixture indicative for weak interaction between unlike molecule leading to increase in molar polarization.

KEYWORDS

Refractive index;
Binary mixture;
Molar polarization;
Average deviation.

INTRODUCTION

Refractive index and density measurement of binary liquid mixture is essential for determination of composition of binary liquid mixture usually for non-ideal mixture where direct experimental measurements are performed over the entire composition range. Most empirical approaches for calculating the excess properties is an attempt to explain non-ideality in term of specific and non-specific intermolecular interactions. The most widely used rules for predictivity refractive in case of binary liquid mixture are Arago-Biot[1], Gladstone-dale[2], Lorentz-Lorenzp[3,4], Eykman[5], Weiner[6], Heller[7], Newton[8], Oster[9] and Eyring-John[10]. Many authors[11-19] have applied these properties to study the structures, solvent-solute interaction and the solvation behavior in binary liquid mixtures.

Oleic acid is a fatty acid found in animal and vegetable oils and occurs naturally in greater quantities than any other fatty acid. High concentrations of Oleic acid can lower blood levels of cholesterol. It is used in the food industry to make synthetic butters and cheeses, flavor baked goods, candy, ice cream, and sodas etc.

EXPERIMENTAL

Oleic acid, methanol, ethanol, n-propanol, n-butanol and iso-butanol (all AR grade products from S. D. Fine Chemicals, India) used in this study were purified by using the methods described in the literature[20,21] Binary mixtures were prepared by mixing of different volumes of two liquids in specially designed ground glass air tight ampoules and weighed in single pan balance (Mettler Toledo AB 204 electronic balance) to an accuracy of ±0.0001gm. Preferential evaporation losses of solvent from the mixture were kept to a minimum as evidenced by repeated measurements of physical properties over an interval of 2-3days during which time no change in physical properties were observed.

The possible error in mole fraction is estimated to be around 0.0001. Densities accurate to $\pm$ 0.00001g$\times$ cm^{-3} were measured by using pycnometer having bulb volume of $10cm^{-3}$ and capillary with internal diameter 1mm for each measurement. The pycnometer was calibrated by using conductivity water (conductivity was $\sim 1\times 10^{-6} ohm^{-1}\times cm^{-1}$). Sufficient time was allowed to attain thermal equilibrium in High Precision Water Bath, Cat No. MSW-274 thermostat, the bath temperature was monitored to $\pm 0.01^0$C with a calibrated thermometer. The refractive index for the sodium D line of the pure components and their mixtures were measured with an Abbe's Refractometer, SER NO. 95033 with an error less than 0.0001 units.

RESULTS AND DISCUSSION

Refraction arises from the presence of extra nuclear electron of atom and tend to follow the oscillations of the electro-magnetic field associated with light. As per Clausius-Mosotti equation at a particular frequency of light $[R]=4\pi N\alpha/3$, while N=Avogadro's number and α is electronic polarizability of the molecules of medium. Polarizability of medium is sum of polarizabilities of constituent atoms. Refractive index depends on the wavelength of light.

In general molar refraction increases with molecular weight for symmetric and asymmetric molecules. Density and refractive index depends on molecular weight and nature of solution. The effect of interaction between the two components become more and more predominant as the alkyl group of the normal alcohols increase in size due to their electron donating ability. Density and refractive index value decrease with increase of temperature from 303.15K to 313.15K.

Molar refraction for the selected binary mixture of oleic acid with alcohols is in the following order:

methanol<ethanol<n-propanol<n-butanol < iso-butanol.

The experimental refractive indices of oleic acid with alcohols are presented in TABLE 1 and the data have been used to evaluate refractive index deviations, via following equation

$$n^E = n_{exp} - \Sigma x_i n_i \quad (1)$$

where x_i and n_i represent the mole fraction and the refractive index of i^{th} component respectively and n_{exp} is the refractive index of the binary liquid mixture .The refractive index devia-

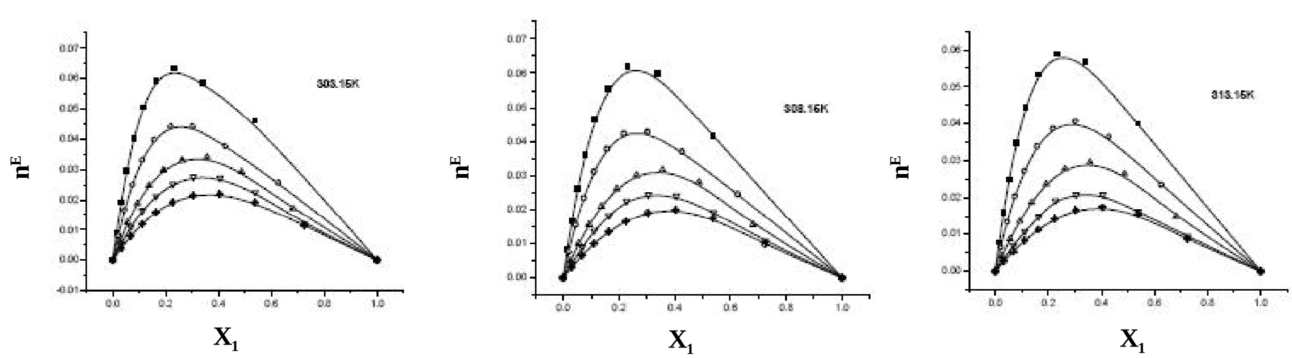

Figure 1: Refractive index deviation for oleic acid (x_1) + methanol (x_2) (■),oleic acid (x_1) + ethanol (x_2) (○),oleic acid (x_1) + n-propanol (x_2) (Δ),oleic acid (x_1) + n-butanol (x_2) (∇), oleic acid (x_1) + iso-butanol (x_2) (◇) at 303.15K, 308.15K and 313.15K

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TABLE 1 : Refractive index density and refractive index deviation of binary system of oleic acid with alcohols at 303.15K, 308.15K and 3313.15K

	Oleic acid (x_1) + methanol (x_2)								
	303.15 k			308.15 K			313.15 k		
X_1	ρ (g cm^{-3})	n_{exp}	n^E	ρ (g cm^{-3})	n_{exp}	n^E	ρ (g cm^{-3})	n_{exp}	n^E
0.0000	0.78194	1.3270	0.0000	0.77772	1.3253	0.0000	0.77237	1.3221	0.0000
0.0141	0.79590	1.3376	0.0088	0.79185	1.3352	0.0081	0.78667	1.3317	0.0078
0.0312	0.80981	1.3501	0.0191	0.80581	1.3459	0.0166	0.80070	1.3420	0.0159
0.0523	0.82334	1.3632	0.0295	0.81940	1.3581	0.0261	0.81450	1.3537	0.0249
0.0791	0.83648	1.3774	0.0402	0.83275	1.3716	0.0362	0.82770	1.3669	0.0347
0.1141	0.84891	1.3922	0.0505	0.84534	1.3863	0.0464	0.84021	1.3810	0.0443
0.1619	0.86033	1.4073	0.0594	0.85698	1.4014	0.0554	0.85166	1.3962	0.0534
0.2311	0.87024	1.4202	0.0634	0.86713	1.4167	0.0619	0.86165	1.4106	0.0589
0.3400	0.87800	1.4295	0.0586	0.87519	1.4286	0.0599	0.86977	1.4223	0.0567
0.5369	0.88298	1.4423	0.0460	0.88066	1.4355	0.0416	0.87626	1.4307	0.0399
1.0000	0.88820	1.4560	0.0000	0.88614	1.4530	0.0000	0.88213	1.4500	0.0000
	Oleic acid (x_1) + ethanol (x_2)								
0.0000	0.78070	1.3586	0.0000	0.77640	1.3562	0.0000	0.77215	1.3532	0.0000
0.0202	0.79460	1.3686	0.0080	0.79075	1.3658	0.0076	0.78663	1.3617	0.0065
0.0443	0.80840	1.3794	0.0165	0.80486	1.3760	0.0155	0.80083	1.3709	0.0134
0.0736	0.82190	1.3908	0.0250	0.81861	1.3868	0.0235	0.81462	1.3806	0.0203
0.1101	0.83520	1.4024	0.0331	0.83185	1.3980	0.0311	0.82784	1.3910	0.0271
0.1565	0.84790	1.4136	0.0398	0.84436	1.4092	0.0379	0.84027	1.4022	0.0339
0.2177	0.85971	1.4240	0.0442	0.85587	1.4195	0.0423	0.85162	1.4130	0.0387
0.3021	0.87010	1.4322	0.0442	0.86600	1.4282	0.0428	0.86155	1.4231	0.0407
0.4259	0.87822	1.4378	0.0377	0.87438	1.4345	0.0371	0.86975	1.4309	0.0365
0.6254	0.88312	1.4452	0.0257	0.88070	1.4413	0.0246	0.87626	1.4372	0.0235
1.0000	0.88820	1.4560	0.0000	0.88614	1.4530	0.0000	0.88213	1.4500	0.0000
	Oleic acid (x_1) + n-propanol (x_2)								
0.0000	0.79559	1.3821	0.0000	0.79140	1.3794	0.0000	0.78739	1.3765	0.0000
0.0257	0.80688	1.3901	0.0061	0.80265	1.3862	0.0049	0.79819	1.3826	0.0042
0.0560	0.81796	1.3987	0.0125	0.81376	1.3936	0.0101	0.80897	1.3894	0.0088
0.0924	0.82073	1.4076	0.0187	0.81641	1.4017	0.0155	0.81130	1.3969	0.0136
0.1367	0.83179	1.4169	0.0247	0.82768	1.4104	0.0209	0.82249	1.4052	0.0187
0.1919	0.84246	1.4261	0.0298	0.83867	1.4196	0.0261	0.83357	1.4142	0.0236
0.2626	0.85268	1.4345	0.0330	0.84925	1.4288	0.0301	0.84440	1.4235	0.0277
0.3565	0.86230	1.4424	0.0340	0.85926	1.4373	0.0317	0.85475	1.4322	0.0295
0.4871	0.87132	1.4473	0.0292	0.86851	1.4433	0.0280	0.86420	1.4387	0.0264
0.6812	0.87990	1.4495	0.0171	0.87696	1.4450	0.0155	0.87242	1.4414	0.0148
1.0000	0.88820	1.4560	0.0000	0.88614	1.4530	0.0000	0.88213	1.4500	0.0000
	Oleic acid (x_1) + n-butanol (x_2)								
0.0000	0.80209	1.3958	0.0000	0.79819	1.3930	0.0000	0.79432	1.3907	0.0000
0.0315	0.81099	1.4032	0.0055	0.80723	1.3990	0.0041	0.80333	1.3958	0.0032
0.0682	0.81983	1.4109	0.0110	0.81622	1.4060	0.0089	0.81230	1.4018	0.0071
0.1115	0.82860	1.4188	0.0163	0.82516	1.4134	0.0137	0.82122	1.4081	0.0108
0.1633	0.83730	1.4268	0.0212	0.83403	1.4210	0.0182	0.83008	1.4151	0.0147
0.2265	0.84592	1.4346	0.0252	0.84284	1.4291	0.0225	0.83887	1.4233	0.0192
0.3052	0.85446	1.4417	0.0275	0.85158	1.4357	0.0244	0.84760	1.4295	0.0207
0.4059	0.86293	1.4474	0.0272	0.86026	1.4414	0.0240	0.85627	1.4355	0.0207
0.5395	0.87135	1.4507	0.0224	0.86888	1.4446	0.0192	0.86489	1.4388	0.0161
0.7249	0.87978	1.4515	0.0121	0.87751	1.4472	0.0107	0.87350	1.4432	0.0095
1.0000	0.88820	1.4560	0.0000	0.88614	1.4530	0.0000	0.88213	1.4500	0.0000
	Oleic acid (x_1) + iso-butanol (x_2)								
0.0000	0.79430	1.3921	0.0000	0.79022	1.3897	0.0000	0.78590	1.3872	0.0000
0.0313	0.80526	1.3980	0.0039	0.80126	1.3948	0.0031	0.79679	1.3918	0.0026
0.0677	0.81590	1.4044	0.0080	0.81202	1.4005	0.0065	0.80745	1.3968	0.0053
0.1107	0.82619	1.4111	0.0119	0.82244	1.4067	0.0100	0.81784	1.4025	0.0083
0.1622	0.83604	1.4183	0.0158	0.83248	1.4134	0.0134	0.82790	1.4087	0.0113
0.2251	0.84544	1.4256	0.0191	0.84209	1.4206	0.0167	0.83760	1.4155	0.0142
0.3035	0.85437	1.4329	0.0214	0.85127	1.4280	0.0191	0.84691	1.4227	0.0164
0.4040	0.86292	1.4398	0.0219	0.86008	1.4352	0.0199	0.85586	1.4299	0.0173
0.5375	0.87130	1.4455	0.0191	0.86871	1.4412	0.0175	0.86458	1.4364	0.0154
0.7233	0.87987	1.4498	0.0115	0.87749	1.4453	0.0098	0.87334	1.4413	0.0087
1.0000	0.88820	1.4560	0.0000	0.88614	1.4530	0.0000	0.88213	1.4500	0.0000

tion were fitted with Redlich-Kister[22] polynomial equation of the from

$$n_E = x_1 x_2 \sum_{i=0}^{k} A_i \,(x_1 - x_2)^i \tag{2}$$

where k is the number of estimated parameters and A_i are the polynomial coefficient evaluated by fitting the equation the experimental with a least square regression method. The standard deviation σ where calculated using the expression

$$\sigma = [\Sigma\,(n_{exp} - n_{cal})^2 / (N\text{-}K)]^{1/2} \tag{3}$$

where N is the number of measurement. The value of coefficient A_i and the standard deviation are presented in TABLE 2

Figure 1 represents the values of refractive index deviation of oleic acid with alcohol varying with the mole fraction of the first component for the binary mixture at 303.15K, 308.15K and 313.15K. n^E value for all system are positive over the complete mole fraction range indicative of increase in polarization may lead to strong intermolecular interaction related to decrease in molar volume and may be a positive enthalpy change on mixing.

Different nine scientists had used following nine equations for quantitative determination of refractive indexes of binary solutions.

Arago-biot (A-B)

$$N = n_1\phi_1 + n_2\phi_2 \tag{4}$$

Dale-gladstone (D-G)

$$n-1 = (n_1-1)\phi_1 + (n_2-1)\phi_2 \tag{5}$$

Lorentz-lorenz (L-L)

$$\frac{n^2-1}{n+0.4} = \left(\frac{n_1^2-1}{n_1+0.4}\right)\phi_1 + \left(\frac{n_2^2-1}{n_2+0.4}\right)\phi_2 \tag{6}$$

Eykman (Eyk)

$$\frac{n^2-1}{n+0.4} = \left(\frac{n_1^2-1}{n_1+0.4}\right)\phi_1 + \left(\frac{n_2^2-1}{n_2+0.4}\right)\phi_2 \tag{7}$$

Weiner (W)

$$\frac{n^2-n_1^2}{n^2+2n_2^2} = \left(\frac{n_2^2-n_1^2}{n_2^2+2n_1^2}\right)\phi_2 \tag{8}$$

Heller (H)

$$\frac{n-n_1}{n_1} = \frac{3}{2}\left[\frac{(n_2/n_1)^2-1}{(n_2/n_1)^2+2}\right]\phi_2 \tag{9}$$

Newton (Nw)

$$n-1 = (n_1^2-1)\phi_1 + (n_2^2-1)\phi_2 \tag{10}$$

Oster (Os)

$$\frac{(n^2-1)-(2n^2+1)}{n^2} = \frac{(n_1^2-1)-(2n_1^2+1)}{n_1^2}\phi_1 + \frac{(n_1^2-1)-(2n_2^2+1)}{n_2^2}\phi_2 \tag{11}$$

Eyring and John (E-J)

$$n = n_1\phi_1^2 + 2(n_1 - n_2)^{1/2}\phi_1\phi_2 + n_2\phi_2^2 \tag{12}$$

In all these equations,

Here, n =Refractive index of mixture, n_1= Refractive index of pure component-1, n_2= Refractive index of pure component-2, ϕ_1=volume fraction of pure component-1, ϕ_2=volume fraction of pure component-2 where,

$$\phi_1 \frac{X_1V_1}{\sum X_iV_i} \;\&\; \phi_2 = \frac{X_2V_2}{\sum X_iV_i} \tag{13}$$

Here, x is the mole fraction v is the molar volume of component.

Deviations with positive sign are significant up to four digit of decimal in all the nine rules so studied. The system can be considered to be nearly ideal one for the rest of various other mixing rules. The APD values for all the systems ranges from 0.0505 to 0.0142. Since liquids of different nature, which has significant differ-

TABLE 2 : Parameters A_i and standard deviation σ for binary liquid mixtures at 303.15K, 308.15K and 313.15K for n^E

T (K)	A_0	A_1	A_2	σ
Oleic acid (x_1) + methanol (x_2)				
303.15	0.1899	-0.0948	0.3983	0.00016
308.15	0.1892	-0.1948	0.2098	0.00019
313.15	0.1729	-0.1825	0.2113	0.00011
Oleic acid (x_1) + ethanol (x_2)				
303.15	0.1315	-0.1299	0.1707	0.00008
308.15	0.1283	-0.1314	0.1431	0.00007
313.15	0.1281	-0.1306	0.0813	0.0001
Oleic acid (x_1) + n-propanol (x_2)				
303.15	0.1141	-0.1099	-0.0305	0.00011
308.15	0.1097	-0.1018	-0.0117	0.00012
313.15	0.1046	-0.0918	-0.0252	0.00011
Oleic acid (x_1) + n-butanol (x_2)				
303.15	0.0966	-0.0831	0.0048	0.00011
308.15	0.0877	-0.0743	-0.0129	0.00011
313.15	0.0761	-0.0605	-0.0217	0.00011
Oleic acid (x_1) + iso-butanol (x_2)				
303.15	0.0807	-0.0524	0.0001	0.00012
308.15	0.0740	-0.0482	-0.0167	0.00012
313.15	0.0651	-0.0396	-0.0191	0.00012

TABLE 3 : Average percentage deviation (APD)α in the refractive index from deferent mixing relations for binary mixtures at various temperatures

	303.15 K	308.15 K	313.15 K
	Oleic acid (x_1) + methanol (x_2)		
A-B	0.0490	0.0467	0.0453
G-D	0.0490	0.0467	0.0453
L-L	0.0491	0.0468	0.0454
W	0.0356	0.0336	0.0321
H	0.0505	0.0482	0.0467
Nw	0.0488	0.0466	0.0451
E-J	0.0491	0.0468	0.0454
Eyk	0.0490	0.0468	0.0453
Os	0.0489	0.0466	0.0452
	Oleic acid (x_1) + ethanol (x_2)		
A-B	0.0374	0.0363	0.0343
G-D	0.0374	0.0363	0.0343
L-L	0.0376	0.0364	0.0344
W	0.0300	0.0289	0.0269
H	0.0383	0.0371	0.0351
Nw	0.0373	0.0361	0.0341
E-J	0.0375	0.0363	0.0344
Eyk	0.0375	0.0363	0.0343
Os	0.0374	0.0362	0.0342
	Oleic acid (x_1) + n-propanol (x_2)		
A-B	0.0284	0.0263	0.0248
G-D	0.0284	0.0263	0.0248
L-L	0.0285	0.0264	0.0250
W	0.0243	0.0222	0.0207
H	0.0289	0.0267	0.0253
Nw	0.0283	0.0261	0.0247
E-J	0.0284	0.0263	0.0249
Eyk	0.0284	0.0263	0.0249
Os	0.0283	0.0262	0.0248
	Oleic acid (x_1) + n-butanol (x_2)		
A-B	0.0230	0.0210	0.0187
G-D	0.0230	0.0210	0.0187
L-L	0.0231	0.0210	0.0188
W	0.0204	0.0183	0.0161
H	0.0233	0.0213	0.0190
Nw	0.0229	0.0209	0.0186
E-J	0.0231	0.0210	0.0187
Eyk	0.0231	0.0210	0.0187
Os	0.0230	0.0209	0.0186
	Oleic acid (x_1) + iso-butanol (x_2)		
A-B	0.0202	0.0186	0.0170
G-D	0.0202	0.0186	0.0170
L-L	0.0203	0.0187	0.0171
W	0.0172	0.0157	0.0142
H	0.0205	0.0189	0.0174
Nw	0.0201	0.0185	0.0169
E-J	0.0202	0.0186	0.0171
Eyk	0.0202	0.0186	0.0171
Os	0.0201	0.0185	0.0170

ence in molecular size, are considered a particular relation provides excellent agreement at some place and deviates at others. The observed deviations are expected and can be ascribed to the volume additively. As during mixing, excess volume is the measurement of molecular interaction in liquid mixtures. The structural property of liquid and liquid mixtures can be integrated through refractive indices employing molar refraction.

A close perusal of TABLE 3 reveals that for all binary mixtures these mixing rules show a good agreement. In all selected systems, refractive index values predicted by Weiner has shown excellent agreement with experimental values followed by Lorentz-Lorenz and Eyring-John relation, which gives fairly good results, where as deviation from theoretical values are more in case of Heller relation. Weiner relation has performed well in case of all binary systems because the variation and deviation with concentration is monotonic without a maximum and without a change in sign. The deviations are within positive values in all the nine rules so studied. In general for binary systems, the deviations are found to be positive where dispersion and dipolar interactions are operating. Some of the factors which may contribute to these deviations are (i) Breaking of hydrogen bonding between alcohol molecules,(ii) Interstitial accommodation of one component into other component, (iii) Possible hydrogen bonding between oleic acid and alcohol molecules and (iv) Interaction between unlike molecules, (v) Dispersive and Dipole interaction. It is suggested that deviation of theoretical values from experimental values can be reduced if excess volumes is taken into consideration. So the small negative deviation can be accounted for the volume deviation without consideration that there is no change in molecular polarizability on mixing of the component.

For all the five binary mixtures, Heller relation has exhibited deviation higher in value than all other relations at all temperatures.

It should be noted that the average percentage deviation value between the Arago-Biot and Gladstone-Dale relations are exactly the same for all system at all temperature. This is expected because of similarities in the function as forms of this equation; as per literature[23] Gladstone-dale relation is more frequently used than the arago-biot equation.

CONCLUSION

All nine mixing rules could be successfully applied at lower concentration of alcohol omitting other factors such as volume reduction, volume addition and temperature. Heller equation could not give better results. All nine theoretical mixing rules performed well within the limits of experiment error. The deviation between theoretical and observed value of refractive indices for all system taken under consideration can be reduced if the concept of excess molar volumes is taken into consideration. Results from Redlich -Kister polynominal equation reveal that more polarizability may lead it strong intermolecular interaction related to decrease in molar volume and positive enthalpy change on mixing.

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